

REMARKS

Restriction Requirement

A restriction requirement has been made between Group I, claims 1-8, and Group II, claims 9-18, because Groups I and II lack the same or corresponding special technical features. A provisional election was made during a telephone conversation on March 28, 2008 to prosecute the invention of Group I, claims 1-8. Applicants hereby confirm the election of Group I, claims 1-8 with traverse. The restriction requirement is traversed on the basis that claims 1-8 and claims 9-18 share at least one general inventive concept of introducing two vapors together before application to a substrate. This feature is provided in both sets of claims: e.g., in claim 1 as “previously mixed” vapors and in claim 9 as vapor streams that “meet each other”.

The present invention is directed to a process for forming a metal oxide film comprising a vapor deposition step in which a vapor of a hydrolysable metal compound and water vapor are brought into contact with a substrate to form a film of a metal oxide precursor on the surface of the substrate and a calcination step in which the substrate is then heated in an oxidizing atmosphere to convert the precursor into a metal oxide. During the vapor deposition step, the hydrolysable metal compound vapor and the water vapor are mixed and the mixed vapors are brought into contact with the substrate within three seconds after mixing. By bringing the hydrolysable metal compound vapor and water vapor into contact with a substrate within three seconds of mixing, the occurrence of a hydrolysis reaction in locations other than at the surface of the substrate can be suppressed. Thus, a significant improvement in the rate of film formation and in the utilization factor of the vapor deposition reactant (metal compound) for film formation is achieved. Additionally, the uniformity of thickness and appearance of the oxide film is improved.

RESPONSE TO REJECTIONS

Claims 1, 5 and 8 are rejected under 35 U.S.C. §103(a) as being unpatentable over U.S. Patent No. 5,919,726 to Hatano et al. in view of U.S. Patent Application Publication No. 2002/0106321 to Tanaka et al.

The Office Action asserts that Hatano et al. teaches a method for producing a photocatalyst material essentially as claimed, except for a three-second vapor mixing time. The Office Action relies on Tanaka et al., directed to the production of ultra-fine particulate titanium oxide particles obtained from titanium tetrachloride by a vapor phase process wherein the titanium tetrachloride vapor and an oxidizing gas are introduced into a reaction chamber with a residence time of three seconds or less. This introduction into the reaction chamber results in the production of particulates, particularly ultra-fine particulate, titanium oxide having a high rutile content in the relationship of BET specific surface area vs. rutile content (see paragraph [0046] of Tanaka et al.). The Office Action then asserts that it would have been obvious to utilize Tanaka et al.'s three-second reaction/mixing time in Hatano et al.'s method in order to ensure the integrity of the titanium oxide film, while preventing clogging of chamber parts as a result of premature product formation within the reaction chamber.

Applicants traverse this rejection for the following reasons.

- **Hatano et al. fails to teach the claimed feature of pre-mixing a vapor of a hydrolysable metal compound and water vapor and bringing the mixed vapors into contact with the substrate within three seconds after mixing.**

Hatano et al. does not teach the vapor deposition defined in claim 1 in which a vapor of a hydrolysable metal compound and water vapor are brought into contact with a substrate to form a film of a metal oxide precursor on the surface of the substrate, and particularly in such a manner that the hydrolysable metal compound vapor and the water vapor are previously mixed before they are brought into contact with a substrate. Hatano et al. actually teaches that water vapor is applied during a first step during silanol application and the titanium tetrachloride is applied in a second step without water vapor. In particular, Hatano et al. teaches that during the coating method, the titanium tetrachloride is mixed with oxygen or dried air and reaction occurs on a coated surface.

In the process disclosed by Hatano et al., a silica gel-containing undercoating, which contains silanol (-Si-OH) groups, is formed in a first step. This undercoating can be formed by coating a substrate with a mixture of silica gel and water (column 2, lines 27-43). In the first step, a water vapor may be positively supplied to the atmosphere so as to form silanol groups sufficiently (column 3, lines 17-20).

In a second step, a titanium tetrachloride vapor is brought into contact with the undercoating having silanol groups, whereby it is vapor-deposited on the undercoating by a condensation reaction with the silanol groups on the undercoating as discussed in column 3, lines 21-22. The contact can be performed by bubbling liquid TiCl_4 with oxygen or dry air and atomizing or spraying the resulting TiCl_4 vapor onto the undercoating. Optionally, a water vapor may be sprayed to recover the excess TiCl_4 vapor as an aqueous solution after or during the supply of the TiCl_4 vapor (column 3, lines 21-58).

Thus, in the process disclosed by Hatano et al., vapor deposition takes place by a condensation reaction between TiCl_4 vapor and silanol groups which have been previously formed on the substrate, and no water vapor is necessary for vapor deposition. A water vapor may be sprayed during or after vapor deposition, but this water vapor is supplied for recovery of the excess or unreacted TiCl_4 by dissolution to form an aqueous solution and does not participate in the vapor deposition reaction (condensation reaction). Such dissolution of TiCl_4 to form an aqueous solution interferes with a vapor deposition reaction. In a CVD technique, unreacted or excess reactant vapors should normally be discharged or expelled from the vapor deposition chamber as they are without condensation.

Accordingly, Hatano et al. fails to teach at least the following features of claim 1

- ❖ a hydrolysable metal compound vapor and a water vapor are previously mixed and the mixed vapors are brought into contact with a substrate for vapor deposition; and
- ❖ the contact of the mixer vapors with substrate occurs within three seconds after mixing.
- **Tanaka relates to production of particulates, not a deposited film**

The Office Action relies on Tanaka et al. as teaching the claimed feature that the hydrolysable metal compound vapor and the water vapor are mixed and brought into contact with the substrate within three seconds after mixing.

Applicants respectfully traverse this position as Tanaka et al. teaches reacting titanium tetrachloride with an oxidizing gas, which may be steam, at a high temperature for a residence time of three seconds or less to obtain particulates. Tanaka et al. is silent with respect to the step of coating a substrate with the mixed metal oxide film, let alone specifically teaching

that the mixed vapors are brought into contact with the substrates within three seconds after mixing.

In particular, Tanaka et al. teaches the production of particulate titanium oxide by reacting a titanium tetrachloride-containing gas and an oxidizing gas, both preheated to 900°C or higher, with a residence time in the temperature range above 700°C which is controlled to be three seconds or less. The oxidizing gas may be steam (water vapor).

The product produced by Tanaka et al. is particulate titanium oxide, rather than a titanium oxide film, as produced by the present invention. In the production of a particulate titanium oxide, the reaction occurs in the gaseous (vapor) phase to form solid fine particles in the absence of a substrate, and all the gases which have reacted in this manner constitute the product. The restriction of three seconds or less in the residence time above 700°C is for the purpose of controlling the growth of particles in order to obtain an ultra-fine particulate product ([0043]).

However, in vapor deposition or CVD technique, to form a titanium oxide film, such a reaction occurring in the vapor phase to form a fine powder is undesirable, since it leads to a wasteful consumption of TiCl_4 vapor (a decrease in film formation rate and in % utilization of TiCl_4 for film formation) and the resulting fine powder may adversely affect the film thickness uniformity and appearance of the titanium oxide film which is formed, as discussed on page 2, lines 10-31 of the present specification.

In the present invention, in order to control such undesirable or premature reaction between TiCl_4 vapor and water vapor occurring in the vapor phase to form a particulate solid, the two vapors are mixed within three seconds before contacting the substrate.

Thus, the reaction which is performed by Tanaka et al. to form a particulate titanium oxide is undesirable in the film formation by a CVD technique according to the present invention, and the problems to be solved by the present invention discussed on page 2 of the specification do not cause any problem in the production of a particulate titanium oxide as disclosed by Tanaka et al.

Of further note, the temperature of the gases used in Tanaka et al. (at least 900°C) is extremely high. One skilled in the art of CVD would not use vapors at such high temperature in the vapor deposition step of claim 1 to form a precursor film on a substrate by a CVD technique.

➤ **There is no rationale to combine the teachings of Hatano and Tanaka**

One having ordinary skill in the art would have no reason to combine the teachings of Tanaka et al. (residence time) for producing a particulate titanium oxide by a reaction in the gaseous phase with the teachings of Hatano et al. for producing a film of titanium oxide by a kind of CVD technique, since the formation of particulate titanium oxide (per Tanaka) by a reaction in the gaseous phase is undesired in producing a film.

Furthermore, even if the teachings of the two references could be combined, the combination does not lead to the subject matter of claim 1. First, Hatano et al. does not teach previous mixture of a titanium tetrachloride vapor and a water vapor as reactants to form a film of a titanium oxide precursor as in claim 1. Second, Tanaka et al. does not teach that mixed vapors of titanium tetrachloride and water vapor (steam) are brought into contact with a substrate within three seconds after mixing. Tanaka et al. merely teaches that the residence time of mixed gases in the temperature range above 700°C is limited to three seconds or less in order to suppress the growth of the formed particles. These teachings have no or little relevance to vapor deposition for film formation.

For the reasons set forth above, a *prima facie* case of obviousness against independent claim 1 has not been established.

Accordingly, it is respectfully requested that the rejection of claims 1, 5 and 8, under 35 U.S.C. §103(a), be withdrawn as Hatano et al., as modified by Tanaka et al., fails to render these claims obvious.

Claim 2 is rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano et al. and Tanaka et al. and further in view of U.S. Patent No. 4,293,326 to Terneu et al.

Terneu et al. is relied upon in the Office Action as teaching a coating process whereby a vapor of a hydrolysable metal compound and water vapor are brought into contact in the vicinity of a traveling substrate via gas streams. Claim 2 depends directly from and adds further limitations to independent claim 1 and is believed to be allowable for at least the reasons discussed hereinabove in connection with independent claim 1. Terneu et al. fails to overcome the deficiencies of the Hatano et al. and Tanaka et al. combination in the rejection of claim 1.

Furthermore, according to claim 2, in order to achieve the previous mixing of the vapors within three seconds before contacting the substrate, two jetted vapor streams meet each other before they reach the substrate. As shown in Fig. 1 of the present application, two lines formed by extensions of the nozzles for making the vapor streams intersect with each other before reaching the substrate. However, the apparatus shown in the sole figure of Ternue et al. is different in this respect. The two nozzles of Ternue (11, 12) are both directed on the glass ribbon (1) and do not meet each other before they reach the glass ribbon (1). Thus, the feature of claim 2 of two vapor streams meeting each other before reaching the substrate is not considered by Ternue et al. There is no reason to modify Ternue et al. to change the directions in which the nozzles (11, 12) are directed, since pre-mixing of the vapors is not relevant to the Ternue et al. process.

For the reasons set forth above, it is respectfully requested that the rejection of claim 2 under 35 U.S.C. §103(a) be withdrawn as any combination of the teachings of Hatano et al., Tanaka et al. and Ternue et al. fails to render this claim obvious.

Claims 3-4 are rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano et al., Tanaka et al., Ternue et al. and further in view of WO 89/00549 to Lindner.

Lindner is relied upon as teaching the use of slit nozzles arranged in opposing relation to and toward each other at an angle for use in a coating process. Claims 3 and 4 depend directly from claim 2 and add further limitations to this claim and independent claim 1 and are believed to be allowable for at least the reasons discussed hereinabove in connection with independent claim 1 and dependent claim 2. Lindner fails to overcome the deficiencies of the Hatano et al., Tanaka et al. and Ternue et al. combination in the rejection of claim 2.

Furthermore, Lindner does not teach that two vapors are previously mixed before contacting a substrate, as defined in claim 1, or that two jetted vapor streams meet each other before reaching the substrate as defined in claim 2. This is apparent from Figs. 9 and 17 of Lindner. The two supply nozzles (10a, 10b) are angled with respect to each other, but they do not deliver vapor streams that meet each other before reaching the substrate (12).

For the reasons set forth above, it is respectfully requested that the rejection of claims 3-4 under 35 U.S.C. §103(a) be withdrawn as the combination of Hatano et al., Tanaka et al., Ternue et al. and Lindner fails to render these claims obvious.

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Claims 6 and 7 are rejected under 35 U.S.C. §103(a) as being unpatentable over Hatano et al. and Tanaka et al. and further in view of U.S. Patent No. 4,261,722 to Novak et al.

Novak et al. is relied upon as teaching a coating process wherein the temperature of the substrate to be coated may be in the range of 150-700°C. Claims 6 and 7 depend directly from claim 5 and add further limitations to this claim and independent claim 1 and are believed to be allowable for at least the reasons discussed hereinabove in connection with independent claim 1 and dependent claim 5. Novak et al. fails to overcome the deficiencies of the Hatano et al. and Tanaka et al. combination in the rejection of claims 1 and 5.

For the reasons set forth above, it is respectfully requested that the rejection of claims 6-7 under 35 U.S.C. §103(a) be withdrawn as the combination of Hatano et al., Tanaka et al. and Novak et al. fails to render these claims obvious.

For all of the foregoing reasons, Applicants believe that claims 1-8 are patentable over the cited prior art and in condition for allowance. Reconsideration of the rejections and allowance of all pending claims 1-8 is respectfully requested.

Respectfully submitted,
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